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USABILITY OF CLAY MIXED RED MUD IN HUNGARIAN BUILDING MATERIAL PRODUCTION INDUSTRY

*Zoltán Sas, János Somlai, Gábor Szeiler, Tibor Kovács

*Corresponding author: ilozas@almos.uni-pannon.hu

Institute of Radiochemistry and Radioecology, University of Pannonia, P.O. Box 158, H-8201 Veszprém, Hungary

Abstract

The Naturally Occurring Radionuclide (Ra-226, Th-232 and K-40) content of building Materials (NORM) contributes to the total radiation dose experienced by humans. In this survey 27 clay and 68 red mud samples were surveyed with gamma spectrometry and screened according to European Basic Safety Standards (BSS) I-index. It was found that average I-index of clays was 0.6 (0.4-0.8) less than the I-index of 1, which makes them suitable for building material production. The average I-index of red mud 2.3 (1.3-3.0). The maximal mixing ratio of red mud was calculated, varied between 12-39 %, with 23 % average.

Keywords: red mud, gamma spectrometry, building material, I-index

Introduction

The building materials found in our homes can have an impact on human health, the indoor environment can greatly depend on the composition of the building materials. The development and application of concepts such as ‘zero waste’, the ‘circular economy’, ‘industrial ecology’ and ‘resource efficiency’ to 21st century economies requires the development of new building materials, this is one reason why reuse of industrial resources (waste streams) becomes a necessity. In case of several processing industries huge amount of by-products are generated, these are often expensive to dispose of but they can be potential raw materials for the construction industry.

One of the most commonly used building materials are bricks made from clay. Due to the congenial internal structure of clays they can be easily mixed with other similar materials without reduction in engineering properties, this provides many possibilities to reuse similar industrial by-products as additive materials in bricks. [1].

The reuse of the residues could reduce the environmental impact of the landfilled by-products, the health risks from dust generation from red mud reservoirs can also be prevented. The integration and correct stabilisation of these by-products into safe building material creates a product with value, and saves money on disposal costs.

The materials with elevated natural radionuclide content (K-40; U-238; Th-232 and their decay products) are called NORM. The exposure pathways have to be identified and quantified in order to manage and remove the potential risks to future residents in buildings containing NORM products [2-4]. In the European Union (EU) the I-index serves as a conservative screening tool to screen of building material, wherein only the gamma exposure is limited.

The production and design of new types of synthetic building materials based on NORM by-products is raising concerns among authorities, public and scientists. It is incumbent on professional engineers and scientists to demonstrate that this material does not pose significant risks to human health and the environment. Although, the reuse of enlisted materials has significant beneficial effect on environment and sustainability the relatively high potential of gamma exposure and elevated indoor radon and thoron levels originated from enhanced Ra-226 and Th-232 content, may increase human health risks [5,6]. Several NORM residues produced in large quantity, such as: phospogypsum (phosphate industry), red mud (aluminum processing industry), fly ash, coal slag (coal burning and steelworks) and so on are presently under investigation [7-17].

The aluminum manufacturing in Hungary started in 1943. As a result of the bauxite refining activities up to now approximately 60 Mt of red mud has been produced in Hungary (30 Mt in Ajka), stored in waste pounds. The Bayer process is the main industrial method to produce alumina (aluminum oxide). During the treatment the bauxite is digested by hot solution of NaOH. This converts the aluminum oxide in the ore to soluble sodium aluminate, $NaAlO_2$. The remaining dissolved particles is clarified by filtering. The remaining mixture of solid residue is called red mud. The natural radionuclide content of the bauxite and resulting red mud, which is increased during Bayer process usually exceeds the world average for soils.

In this study 68 red mud samples were collected from eight red mud reservoirs of Ajka (Hungary). 27 clay samples were taken, these are used in 14 Hungarian building material factories (across every region of the country). The function of the samples radionuclide content determined the permissible mixing ratio, which was calculated according to the I-index recommended by BSS [18].

MEASURMENTS AND METHODS

Sample collection

During this survey 68 red mud samples were collected from 1-2 m depth of “I”, “II”, “IV”, “VI”, “VII”, “VIII”, “IX” and “X” red mud reservoir of Ajka (Hungary). 14 samples from brick producers were analysed and baseline samples were gathered from 27 different clayfields to screen the natural radioactivity content of clay soils.
The location of these samples are illustrated in Figure 1.

Fig.1 Sampling locations

Determination of Ra-226, Th-232, K-40 activity concentration by gamma-spectrometry

The samples were dried to constant mass in oven at temperature of 105 °C. Before they were stored for 30 d in air-tight aluminum Marinelli beakers with volume of 600 cm^3 to reach the secular equilibrium between the Ra-226 and the Rn-222 (and their decay products) the samples were crushed in mortar and sieved under 0.63 mm mesh diameter.
The determination of the Ra-226 activity concentration occurred via the radon progenies Pb-214 (295 keV) and Bi-214 (609 keV), the Th-232 content was obtained from Ac-228 (911 keV and Tl-208 (2614 keV) and the K-40 with the help of 1460 keV gamma line by high resolution gamma ray spectrometry, using an ORTEC GMX40-76 HPGe semiconductor detector with efficiency of 40 %, and an energy resolution of 1.95 keV at 1332.5 keV.

The data and spectra recorded by a Tennelec PCA-MR 8196 MCA. The system was calibrated with IAEAsoil reference material. The sample measuring time was 80.000 s in every case.

Screening samples with I-index

The European Commission (EC) set guidelines on the radiological protection principles concerning the natural radioactivity of building materials (RP-112 document) for the Member States [17].

An activity index is proposed to correspond to the excess gamma exposure (compared to background received outdoors). Doses to members of the public should be kept as low as reasonably achievable. Within the EU, gamma doses exceeding 1 mSv/y should be considered by a competent authority from the radiation protection point of view. The following activity concentration index (I) is derived to identify whether a dose criterion is met:

$$I = \frac{C_{Ra-226}}{300 Bq/kg} + \frac{C_{Th-232}}{200 Bq/kg} + \frac{C_{K-40}}{3000 Bq/kg} \qquad (1)$$

Where C_{Ra-226}, C_{Th-232}, C_{K-40} are the Ra-226, Th-232 and K-40 activity concentrations (Bq/kg) in the building material.

For effective application of the index to residues from industries processing NORMs recycled into building material, an appropriate partitioning factor needs to be applied. The activity concentration index value of 1.0 can be used as a conservative screening tool for identifying materials that may cause the reference level laid down in Article 75(1) of 2013/59/EURATOM council directive to be exceeded [18]. The Council directive allows the dilution and mixing of construction materials as long as the final building product itself (the bricks) are below the activity concentration index value of 1.0.

RESULT AND DISCUSSION

The radionuclide concentrations of Hungarian clay samples are shown in Table 1. The mean Ra-226 activity concentration was 37 ± 7 Bq/kg (16 ± 3 to 105 ± 17 Bq/kg), the mean of Th-232 was 40 ± 9 Bq/kg (31 ± 7 to 49 ± 11 Bq/kg) and the average of the K-40 content was 803 ± 98 Bq/kg (534 ±16 to 1127 ± 105 Bq/kg). The mean values of Hungarian clays are comparable with world average mean radionuclide concentration of soils reported in UNSCEAR 2008 Annex B (Ra-226: 32 Bq/kg, Th-232: 45 Bq/kg, K-40: 412 Bq/kg) and RP112 (Ra-226: 40 Bq/kg, Th-232: 40 Bq/kg, K-40: 400 Bq/kg) [19]. Only the K-40 content was higher than the UNSCEAR and RP112 reference value in every samples [17]. This is due the natural enriched metal content of the soils and bedrock that provide raw materials for the local alumina and metal industries.

The calculated I-index (derived from the natural radionuclide content) of the examined 27 clay samples – used in building material factories – was lower than 1.0 value (average 0.59). Due to that fact it can be stated, that the investigated clays are suitable for bulk amount used building material from gamma dose point of view. The natural radionuclide content of the red mud samples are presented in Table 2. The obtained results clearly prove that the natural radionuclide content of the deposited red mud by product varies in great range, which requires frequent sampling and homogenizing before reuse. The inhomogeneity can be caused by the condition of the processed bauxite residue and deposition factors such as particle size and particle density within the reservoirs.

Table 1 Activity concentrations and I-indexes of investigated clays

ID	Activity Concentration [Bq/kg]			I-index
	Ra-226	Th-232	K-40	
BSZGY I	32 ± 6	34 ± 7	726 ± 32	0.5
BSZGY II	55 ± 9	48 ± 10	1113 ± 55	0.8
BSZGY III	37 ± 7	32 ± 7	690 ± 37	0.5
BSZ I	105 ± 17	42 ± 9	743 ± 105	0.8
BSZ II	22 ± 4	36 ± 8	753 ± 22	0.5
SMR I	30 ± 7	38 ± 10	787 ± 30	0.6
SMR II	30 ± 5	38 ± 8	852 ± 30	0.6
KB	33 ± 5	42 ± 8	885 ± 33	0.6
KSZ	37 ± 7	49 ± 11	731 ± 37	0.6
SP	44 ± 7	44 ± 8	1127 ± 44	0.7
SMB	85 ± 15	39 ± 9	832 ± 85	0.8
TSZV	36 ± 6	43 ± 9	919 ± 36	0.6
BCS	29 ± 6	38 ± 10	856 ± 29	0.6

OBTY	26 ± 6	32 ± 9	760 ± 26	0.5
MZT	31 ± 6	40 ± 10	768 ± 31	0.6
ABNY	30 ± 6	44 ± 10	917 ± 30	0.6
DEVK	38 ± 6	48 ± 8	993 ± 38	0.7
DEVS	32 ± 5	43 ± 7	755 ± 32	0.6
TKV	40 ± 8	40 ± 11	907 ± 40	0.6
BKS I	32 ± 5	38 ± 7	672 ± 32	0.5
BKS II	29 ± 6	43 ± 10	786 ± 29	0.6
MD-PALA	26 ± 5	36 ± 8	751 ± 26	0.5
MD-SAR	16 ± 3	31 ± 7	582 ± 16	0.4
MD-FEK	26 ± 5	36 ± 7	751 ± 26	0.5
MD-MAR	38 ± 6	42 ± 9	813 ± 38	0.6
MD-PIR	23 ± 5	44 ± 10	534 ± 23	0.5
MD-BAR	30 ± 7	39 ± 8	687 ± 30	0.5
AVG	37 ± 7	40 ± 9	803 ± 37	0.6
Min	16 ± 3	31 ± 7	534 ± 16	0.4
Max	105 ± 17	49 ± 11	1127 ± 105	0.8

Table 2 Activity concentrations and I-indexes of investigated red mud samples

ID	Activity Concentration [Bq/kg]			I-index
	Ra-226	Th-232	K-40	
Reservoir I (3 samples)				
AVG	265 ± 29	268 ± 28	20 ± 2	2.2
Min	232 ± 28	256 ± 26	17 ± 2	2.1
Max	298 ± 36	284 ± 28	25 ± 3	2.4
Reservoir II (6 samples)				
AVG	315 ± 33	286 ± 25	29 ± 3	2.5
Min	282 ± 34	258 ± 28	5 ± 1	2.4
Max	349 ± 42	281 ± 25	36 ± 3	2.5

Reservoir IV (3 samples)				
AVG	313 ± 32	276 ± 28	40 ± 4	2.4
Min	275 ± 30	255 ± 26	22 ± 3	2.4
Max	335 ± 37	295 ± 33	56 ± 3	2.5
Reservoir VI (6 samples)				
AVG	332 ± 35	251 ± 24	52 ± 5	2.4
Min	301 ± 24	238 ± 19	28 ± 2	2.3
Max	345 ± 28	262 ± 21	79 ± 5	2.4
Reservoir VII (17 samples)				
AVG	325 ± 31	246 ± 24	50 ± 6	2.3
Min	330 ± 26	236 ± 19	32 ± 3	2.3
Max	372 ± 30	267 ± 24	37 ± 6	2.6
Reservoir VIII (10 samples)				
AVG	348 ± 34	255 ± 25	52 ± 5	2.5
Min	291 ± 23	263 ± 32	41 ± 3	2.3
Max	395 ± 34	273 ± 27	82 ± 9	2.7
Reservoir IX (5 samples)				
AVG	348 ± 35	261 ± 26	55 ± 5	2.5
Min	356 ± 32	247 ± 25	41 ± 3	2.4
Max	435 ± 44	314 ± 31	67 ± 4	3.0
Reservoir X (18 samples)				
AVG	182 ± 17	247 ± 26	284 ± 29	2.0
Min	152 ± 15	129 ± 12	285 ± 29	1.3
Max	215 ± 24	192 ± 26	360 ± 40	1.8
Summary (68 samples)				
AVG	289 ± 31	255 ± 25	110 ± 12	2.3
Min	152 ± 15	129 ± 12	17 ± 29	1.3
Max	435 ± 24	314 ± 26	360 ± 40	3.0

Determination of mixing ratio

In function of the radionuclide content the mixing proportion of red mud can be calculated according to I-index, recommended by RP 112 guideline and EU BSS [18]. On the basis of the obtained results the maximum red mud (from all the investigated reservoirs) mixing ratios were calculated (Table 3.) with all of the investigated Hungarian clays used in building material factories.

Table 3 Mixing ratios of red mud and Hungarian clay samples

Clay ID	Maximum red mud content [%]							
	I	II	IV	VI	VII	VIII	IX	X
BSZGY I	28	24	25	26	27	25	25	34
BSZGY II	15	12	13	13	14	13	12	18
BSZGY III	28	25	25	26	27	25	25	34
BSZ I	14	12	12	12	13	12	12	17
BSZ II	29	25	26	27	27	26	25	35
SMR I	27	23	24	25	25	24	23	32
SMR II	26	22	23	24	24	23	22	31
KB	24	21	21	22	23	21	21	29
KSZ	24	21	21	22	23	21	21	29
SP	17	15	15	16	16	15	15	22
SMB	16	14	14	15	15	14	14	20
TSZV	23	19	20	21	21	20	20	28
BCS	26	22	23	24	24	23	22	31
OBTY	29	25	26	27	27	26	25	35
MZT	26	23	23	24	25	23	23	32
ABNY	24	20	21	22	22	21	20	29
DEVK	20	17	17	18	18	17	17	24
DEVS	26	22	23	24	24	23	22	31
TKV	23	20	20	21	21	20	20	28
BKS I	28	24	25	26	27	25	25	34

BKS II	26	22	23	24	24	23	22	31
MD-PALA	28	25	25	26	27	25	25	34
MD-SAR	33	29	29	30	31	29	29	39
MD-FEK	28	25	25	26	27	25	25	34
MD-MAR	24	21	21	22	23	21	21	29
MD-PIR	30	26	27	28	28	27	26	36
MD-BAR	28	24	25	26	26	25	24	34
Min	14	12	12	12	13	12	12	17
Max	33	29	29	30	31	29	29	39
AVG	25	21	22	23	23	22	22	30

The obtained mixing ratios allow on an average 23 % red mud content in the case of red mud samples. The obtained minimum mixing proportion of red mud was 12 % in the case of sample “BSZ I” and the maximum was 39 % in the case of “BSZ II”. Further work is required to determine the optimal engineering properties of the red mud bricks that are mixed to within an acceptable I value of less than 1.

CONCLUSION

The surveyed clay samples used in Hungarian building material factories have lower I-index (on an average 0.6) than the 1.0 value, which proves, all of them can be used directly as bulk building materials. It can be stated that all of the investigated red mud samples have a higher I-index than 1.0 index value, which is recommended by RP 112 guideline and new EU BSS [18].This requires that if the red mud is to be used it must be mixed so that any final product has an I-index of less than 1. The maximum mixing ratios of the red mud samples and the investigated clay samples were calculated. The mixing proportions of the red mud varied between 12 to 39 % on average of 23 %, which make the economic reuse of mud possible in significant amount of the samples. However, the I-index provide opportunity for screening building materials in general in case of outdoor usage e.g. roof tiles or paving the 1.0 I-index value is not reasoned [20]. Other categories of indexes were defined for outdoor usage but in case of outdoor used building materials further category should be established.[21]

Owing to the inhomogeneity the reuse of red mud as add material might be difficult, since continuous monitoring should be applied in building material factories before mixing to keep the recommended I-index. A monitoring protocol for red mud used in building products should be developed using sampling methods familiar to the ore processing and environmental forensic industries [22,23].

ACKNOWLEDGEMENT

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Max	395 ± 34	273 ± 27	82 ± 9	2.7
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Table 3 Mixing ratios of red mud and Hungarian clay samples

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BSZGY III	28	25	25	26	27	25	25	34
BSZ I	14	12	12	12	13	12	12	17
BSZ II	29	25	26	27	27	26	25	35
SMR I	27	23	24	25	25	24	23	32
SMR II	26	22	23	24	24	23	22	31
KB	24	21	21	22	23	21	21	29
KSZ	24	21	21	22	23	21	21	29
SP	17	15	15	16	16	15	15	22
SMB	16	14	14	15	15	14	14	20
TSZV	23	19	20	21	21	20	20	28
BCS	26	22	23	24	24	23	22	31
OBTY	29	25	26	27	27	26	25	35
MZT	26	23	23	24	25	23	23	32
ABNY	24	20	21	22	22	21	20	29
DEVK	20	17	17	18	18	17	17	24
DEVS	26	22	23	24	24	23	22	31
TKV	23	20	20	21	21	20	20	28
BKS I	28	24	25	26	27	25	25	34
BKS II	26	22	23	24	24	23	22	31
MD-PALA	28	25	25	26	27	25	25	34
MD-SAR	33	29	29	30	31	29	29	39
MD-FEK	28	25	25	26	27	25	25	34
MD-MAR	24	21	21	22	23	21	21	29
MD-PIR	30	26	27	28	28	27	26	36
MD-BAR	28	24	25	26	26	25	24	34

Min	14	12	12	12	13	12	12	17
Max	33	29	29	30	31	29	29	39
AVG	25	21	22	23	23	22	22	30